Fog processing of atmospheric organic matter

P. Herckes[1,2], S. Youngster[1], T. Lee[1] and J.L. Collett[1#]

[1] Atmospheric Science Department, Colorado State University, Fort Collins, CO

[2] Present address: Department of Chemistry, Arizona State University. Tempe, AZ

[#] Corresponding author

Abstract

The scavenging of carbonaceous particulate matter by radiation fogs was investigated at two locations in California's Central Valley (Angiola and Fresno). Concentrations of carbon and select molecular marker species were determined in ambient and interstitial particles and their scavenging efficiencies by the fogs calculated. Results show that fogs in the region can process carbonaceous particles efficiently as total carbon scavenging efficiencies of up to 85% were observed. Organic carbon was scavenged preferentially over elemental or black carbon, resulting in shorter atmospheric lifetimes for the former compared to the latter. Significant differences were observed in the processing of organic molecules utilized as tracers for different source types. Levoglucosan and other wood smoke markers were scavenged very efficiently, while hopanes, indicative of vehicle exhaust, remained mainly in the interstitial (unscavenged) particles. These results suggest that fogs, important cleanser of the atmosphere, discriminate between OC of different sources, favoring scavenging and hence removal of wood smoke over other source types including vehicle emissions.

Keywords: fog chemistry, cloud chemistry, aerosol cloud interaction, scavenging efficiency

1. Introduction

Cloud and fog droplets influence atmospheric composition through scavenging of particles and gases. Scavenged species can undergo transformation in cloud and fog drops and be removed through incorporation into rain/snow and/or direct droplet deposition. Wet and occult deposition processes relevant to inorganic sulfur and nitrogen species have been extensively studied (e.g. Collett et al., 2001; Herckes et al., 2002a). The deposition rates are typically far higher than for dry deposition. Consequently, chemical species that are efficiently scavenged by cloud/fog drops should experience significantly shorter lifetimes than species which are not as efficiently incorporated into atmospheric droplets.

Because past scavenging and deposition studies have focused mainly on inorganic species, relatively little is known about fog/cloud scavenging and deposition of organic species. Recent studies have demonstrated, however, that organic matter is an important component of fog droplets (Anastasio et al., 1994; Fuzzi and Zappoli, 1996; Gelencser et al., 2000; Zhang and Anastasio 2001; Herckes et al., 2002b; Loeflund et al., 2002) and laboratory studies showed that organic species can act very efficiently as cloud condensation nuclei (Cruz and Pandis, 1997; Corrigan and Novakov, 1999; Yu, 2000; Prenni et al., 2001; Prenni et al., 2003). Furthermore, soluble organic gases are readily scavenged by droplets (Laj et al., 1997; Voisin et al., 2000; Ervens et al., 2003). Only a few field studies have addressed scavenging of carbonaceous particles or individual organic species by clouds or fogs (e.g. Facchini et al., 1992; Facchini et al., 1999; Limbeck and Puxbaum, 2000; Hitzenberger et al., 2001). Observations of individual organic species scavenging are often limited to organic acids.

While all studies of bulk carbonaceous aerosol scavenging have found a significant fraction of carbonaceous particulate matter is scavenged by clouds, there is some dissent about the scavenging efficiency of black/elemental carbon (Sellegri et al., 2003). Many studies of cloud/fog scavenging rely on conversion of aqueous cloud water concentrations to air concentrations using a liquid water content measurement or estimate, introducing additional uncertainty into calculated efficiencies. Other studies have made use of virtual impactor measurements, complicating interpretation of scavenging for volatile species, important constituents of atmospheric fog and cloud droplet organic matter. Finally the complexity of the physicochemical processes involved in fog and cloud systems, including relatively fast dynamic changes, including entrainment, compared to long measurement integration times, make fog scavenging experiments quite challenging.

The present study aims to add to our still primitive understanding of carbonaceous particle scavenging by presenting observations of particulate carbon and individual organic species scavenging by California radiation fogs. This effort, in contrast to most previous work, determines efficiencies based solely on observations of particulate matter concentrations, ambient and interstitial, and does not rely on conversion of aqueous phase concentrations to equivalent air concentrations.

2. Experimental

Two fog field studies were conducted in the Central Valley of California. A first study was conducted in winter 2000/01 in Angiola, CA, within the framework of the California Regional PM10/PM2.5 Air Quality Study CRPAQS (e.g. Herckes et al., 2002b; Herckes et al., submitted). A second study was conducted in Fresno, CA in winter 2003/04. While the Angiola site was a remote (but polluted) agricultural location, the Fresno site was located within the city. Sampling was performed on the experimental farm of Fresno State University relatively close (100s of m) to major highways and residential areas. For both studies, cloud samples were collected with a stainless steel version of the Caltech Active Strand Cloudwater Collector (ss-CASCC) allowing for collection of fog droplets larger than 3.5 µm in diameter by impaction on stainless steel strings (e.g., Herckes et al., 2002b).

Aerosol samples were collected on pre-fired quartz fiber filters. In the Angiola study, samples were collected by a 2 channel medium volume (120 $Lmin^{-1}$) aerosol collector (e.g. Brown et al., 2002). In the absence of fog, ambient aerosol samples were collected on the ambient channel, downstream of a $PM_{2.5}$ cyclone. When fog appeared, sampling was manually switched to the second channel where the inlet was situated inside the cloudwater collector downstream of the impaction strings. For the Fresno study, aerosol samples were collected by a high volume sampler (1.13 m^3 min^{-1}, ThermoAndersen, Smyrna, GA) with a PM2.5 impactor inlet (Tisch Environmental TE231). In the absence of fog ambient aerosol was collected; in the presence of fog the aerosol sampler including the PM 2.5 impaction stage were sampling downstream of the collection strands of the ss-CASCC. Collected filter samples were stored frozen until analysis.

Total, Organic and Elemental Carbon (TC, OC and EC) were measured by the thermal optical transmission method (Birch and Cary, 1996). Some of the filters were analyzed by Sunset Laboratories while a subset of filters was analyzed in our laboratory at Colorado State University using a Sunset Instruments semi-continuous carbon analyzer in offline mode. Molecular marker concentrations on filter and fog samples were determined by gas chromatography coupled to mass spectrometry following extraction with dichloromethane. The detailed procedures are described elsewhere (Brown et al., 2002; Herckes et al., 2002c).

In the Fresno study in addition to the filter sampling, black carbon (BC) was determined continuously by a 2-channel aethalometer (Hansen et al., 1984) operated with a stainless steel CASCC2 as an inlet. The 3.5 µm size cut of the CASCC2 (Demoz et al., 1996) prevented most fog drops from reaching the aethalometer when fog was present. Total carbon was determined with a 1 hour time resolution by an R&P continuous particulate carbon monitor (Model 5400). Air sampled by this instrument was drawn first through a CASCC2 and then through a PM2.5 cyclone. TC, OC and EC were determined semi-continuously (1 hr) with a Sunset Instrument semi-continuous analyzer. Air sampled by this instrument was drawn through a stainless steel CASCC2, a $PM_{2.5}$ inlet and a denuder.

Cloud liquid water content was measured at 1 min time resolution by a Gerber PVM 100 (Gerber, 1991).

3. Results and discussion

Total carbon TC scavenging

As a first step we consider the scavenging of total carbon. Figure 1 illustrates a typical evolution of total particulate carbon concentrations (TC). In the absence of fog (indicated by low liquid water content (LWC)) the TC value represents the ambient particulate matter concentration of TC; during a fog event (high LWC) the TC values correspond to interstitial aerosol particles that have not been scavenged by fog drops (by nucleation or other mechanisms). We observe that interstitial TC concentrations are lower than ambient TC concentrations, suggesting that a portion of the fine particle TC was scavenged by fog drops. For example, interstitial aerosol concentrations on 12/18 and 12/19 are significantly lower than ambient fine particle TC concentrations observed in non-foggy periods on the same days. The suggestion of fine particle TC scavenging is supported by high concentrations of total organic carbon (TOC) observed in fogwater collected in these events, although one must keep in mind that fog TOC represents the sum of scavenged particulate OC and gaseous volatile organic compounds (e.g., acetic acid, formic acid, formaldehyde, and other compounds). Speciation of the fog TOC indicated that a large fraction of the fog organic matter most likely originated from OC particle scavenging (Herckes et al., 2002b). It is noteworthy that while figure 1 depicts a general pattern indicative of fog scavenging of TC, a few periods showed TC concentrations that did not decrease as expected. These may reflect changes occurring in atmospheric composition during the sampling periods and/or effects of entrainment from above the boundary layer during fog growth.

Interestingly, post-fog carbonaceous fine particle concentrations are somewhat higher than pre-fog concentrations, raising a question as to whether aqueous reactions in the fog drops might transform soluble VOCs into lower volatility secondary organic aerosol (SOA) species, analogous to aqueous phase transformation of gaseous sulfur dioxide to particulate sulfate. Although transformations of this type have been predicted in the literature (e.g., Blando and Turpin, 2000), the organic chemistry of the atmospheric aqueous phase is largely unknown and the importance of aqueous phase SOA production is mostly speculation at present. Significant aqueous SOA production has not been clearly documented in any field experiment, in part because of the difficulty of adequately characterizing such a complex, multiphase system. Measurements during CRPAQS are also inadequate to do more than speculate whether the presence of the fogs contributed to SOA formation or whether increases in particulate TC concentrations resulted from other mechanisms such as advection or entrainment.

It is difficult to schedule short-term aerosol measurements to correspond exactly to the desired periods before, during, and after fog especially given the rather unpredictable nature of fog onset and dissipation. Interpreting differences between pre- and post-fog samples as fog scavenging and deposition of aerosol carbon is also challenging as the depth of the boundary layer typically grows with onset of the fog, entraining material of unknown concentration and composition from above. Overall, however, our data suggest that elemental carbon (EC) is less efficiently scavenged than OC resulting in decreased OC/EC ratios in the interstitial samples. This is particularly obvious for the Angiola December 18th and 19th period (Figure 1). In this case the ratio OC/EC decreased in the interstitial sample compared to pre-fog conditions and increased again after the fog dissipated, drying out the droplets which become particles again.

Preferential scavenging of OC vs EC is further supported by the data presented in figure 2. Here fine particle EC is plotted vs OC at the Angiola site during periods of intensive $PM_{2.5}$ aerosol monitoring by Desert Research Institute scientists. OC and EC in these filter samples were measured using the thermal optical reflectance (TOR) method. Data are divided into periods with and without fog. Many of the lowest OC/EC ratios occur during periods with fog, consistent with the preferential fog scavenging of OC outlined above. The data are, however, quite scattered and a possible artifact may result from the fact that foggy periods occur preferentially at night. Diurnal variations in OC/EC ratios could bias the results, although the distance of the Angiola site from primary emissions sources should mitigate this effect.

Scavenging efficiencies

For periods with a rapid onset of fog, well represented by ambient and interstitial filter samples, we were able to calculate scavenging efficiencies according to:

$$\eta = 1 - \frac{X_{interstitial}}{X_{pre-fog}}$$

where η is the scavenging efficiency of species X.

It is noteworthy that the expression relies only on particulate concentrations and not on estimates of fog LWC. The results are given in Table 1 for the events where fog scavenging appeared to dominate concentration changes (i.e., TC was observed to decrease upon fog formation). Results are presented for both the Angiola and Fresno fog campaigns. Scavenging efficiencies for OC were calculated to vary between 33 and 90%. Scavenging efficiencies for EC were much lower, ranging from 5 to 12%, again suggesting a higher efficiency for OC scavenging than for EC scavenging. For the Fresno samples a collocated R&P 5400 semi-continuous carbon analyzer yielded a scavenging efficiency of 46% for total carbon in reasonable agreement with the 36% for the Thermal Optical Transmission method. On the other hand, we obtained a higher scavenging for Black Carbon as measured by the aethalometer than for Elemental carbon EC. Nevertheless, both efficiencies are low. Further, EC and BC are different parameters and in wintertime Fresno where ambient particulate matter is strongly influenced by domestic wood burning, the difference might be amplified.

Overall the data show again that EC is scavenged less favorably than OC. The preferential scavenging of OC can be explained by the more hygroscopic character of organic carbon-containing particles compared to particles rich in EC (e.g., soot) and hence a higher likelihood of becoming activated. Some difference in nucleation scavenging efficiency may also result from different size distributions for EC and OC.

The observed differences in scavenging efficiencies have important consequences for particle lifetime, since these radiation fogs have been shown to be effective cleansers of the atmosphere. Deposition velocities of both inorganic and organic solutes contained in fog droplets are much higher than dry deposition velocities for the same species (Collett et al., 2001; Herckes et al., 2002a). The higher fog scavenging efficiencies observed for OC suggest that the atmospheric lifetime of OC in the Central Valley will be limited much more strongly by the occurrence of fogs than will the lifetime of EC. In a somewhat analogous situation, Lim and coworkers observed a shorter lifetime of organic carbon over the ocean during ACE Asia and hypothesized that a contributing factor might be preferential scavenging of OC by clouds (Lim et al., 2003)

Our observed scavenging efficiencies are somewhat similar to previous observations. Hallberg and coworkers measured a scavenging efficiency of 6% for BC in polluted radiation fogs in the Po Valley (Hallberg et al., 1992). Hitzenberger, by contrast, observed much higher scavenging efficiencies for BC in clouds formed in more pristine areas (Hitzenberger et al., 2000; Hitzenberger et al., 2001) where particle scavenging is likely to be enhanced by generation of greater peak supersaturations. Hitzenberger's observed TC scavenging efficiencies, however, were similar in magnitude to those observed in the current study. Surprisingly, Sellegri and coworkers found higher EC than OC scavenging efficiencies. They hypothesized that EC was largely associated with particles rich in other hydrophilic species while OC-containing particles featured hydrophobic coatings that could suppress activation (Sellegri et al., 2003). A similar explanation was presented by Hitzenberger to account for very high BC scavenging.

Figure 3 depicts the variation in TC scavenging efficiency determined in the current study with fog LWC. One observes that efficiencies tend to increase with increasing LWC. Such an increase might reflect correlation of both scavenging efficiency and of fog LWC with peak fog supersaturation (which cannot be measured directly). A linkage of this type is certainly plausible although, given the limited number of observations available here, we should be hesitant in interpreting this relationship too strongly.

Other authors have previously suggested a relationship between LWC and scavenging efficiencies (Hitzenberger et al., 2000; Hitzenberger et al., 2001); such a relationship was not observed, however, by Sellegri and coworkers (Sellegri et al., 2003). One criticism of Hitzenberger's work was that LWC was necessary to convert aqueous concentrations into air concentrations for scavenging efficiency calculations, introducing a strong sensitivity of calculated scavenging efficiencies to LWC. Because LWC was not needed to determine scavenging efficiencies in the current study, the observed relation between efficiency and LWC does not suffer from the same bias.

As described above, the ability to make fog scavenging measurements of this type is limited in part by concentration changes that can occur during the long sampling intervals needed for filter-based sampling. We hoped to minimize this problem in the Fresno field campaign by making use of semi-continuous carbon measurements. Unfortunately EC concentrations were low enough that OC/EC ratios were quite uncertain. In future experiments we hope to make use of even faster, more sensitive measurements of aerosol composition with an aerosol mass spectrometer to continue to address difficulties in accurately determining fog and cloud scavenging efficiencies.

Molecular markers

The results presented thus far suggest that the studied fogs scavenged OC more efficiently than EC. We have previously reported that many organic molecular marker compounds are observed in fog samples (Herckes et al., 2002c), indicating that there is some efficiency of the fogs for scavenging particles from the corresponding source types (e.g., wood smoke, meat cooking, and vehicle exhaust). Examination of the relative scavenging efficiencies of markers from different carbonaceous particle source types can help us determine whether fogs in the region process carbonaceous particles from some source types more actively than others. Such an observation could have important implications for understanding relative atmospheric lifetimes of different carbonaceous particle types during the foggy periods that accompany winter stagnation episodes in California's Central Valley.

We will focus on two fog episodes during which fog scavenging appeared to dominate concentration changes. A first episode occurred in Angiola on December 18/19 2000. This event is extensively discussed in a modeling study (Fahey et al., 2005 in press). The second case study occurred in Fresno on January 11th 2004. Table 2 presents the scavenging efficiency of select individual organic compounds as calculated for both events.

We note, on one hand, that there are significant differences in scavenging efficiencies between species. On the other hand individual marker scavenging efficiencies appear similar in both events. Due to short sampling integration times and relatively low aerosol sampler flows (for CRPAQS), some molecular marker species were close to or below detection limit and scavenging efficiencies could not be determined.

The lowest scavenging efficiencies are observed for non-polar species, including n-alkanes and hopanes; polar species like dicarboxylic acids and levoglucosan showed very high scavenging efficiencies. For n-alkanes, which were present at low concentrations, the scavenging efficiencies averaged 60%. It is noteworthy that there is no significant difference between the alkane carbon preference index (CPI, indicating the relative proportions of alkanes with odd and even carbon numbers) of the interstitial and pre-fog aerosol. n-alkanoic acids, with their single hydrophilic carboxylic acid group show a significantly higher scavenging efficiency than the n-alkanes. Polyaromatic hydrocarbons (PAH) show low scavenging efficiencies, whereas oxy-PAH are scavenged much more efficiently. These results are consistent with observations by Limbeck and Puxbaum (2000) that there is a relationship between scavenging and compound solubility. As shown in previous work, a significant part of the scavenged organic material can be contained in an insoluble phase inside the cloud or fog droplets (Herckes et al., 2002c).

For the Angiola fog event we see that some species are scavenged more efficiently than bulk organic carbon while others are scavenged less efficiently. For the second fog event (Fresno) it appears that all species are scavenged as well or better than bulk OC, although there is again a large difference between individual species.

Levoglucosan, a popular molecular marker for biomass burning (e.g. Simoneit et al., 1999), exhibited very high scavenging efficiencies and was essentially absent from the interstitial particulate matter. This is consistent with high levoglucosan concentrations observed in simultaneously collected fog water, where it is a major organic component. High scavenging efficiencies were also observed for other wood smoke markers, including vanillin and syringaldehyde, while retene appeared to be scavenged less efficiently. Overall wood smoke marker molecules and, hence, smoke particles emitted from biomass combustion are more efficiently scavenged than organic carbon in general. By contrast, 17α21β hopane, a species frequently used as molecular tracer for vehicle emissions (e.g. Schauer et al., 1996) shows a low scavenging efficiency. These observations show that fogs (or clouds) cleanse the atmosphere of particles from select sources (e.g. wood smoke) faster than others (e.g., vehicle emissions). In areas like California's Central Valley, where winter stagnation episodes are frequently accompanied by dense fog formation, it may be worth considering the differential impact that the fogs appear to exert on scavenging and removal of particles from different source types as part of pollution reduction strategies.

4. Summary and conclusions

Total (TC), organic (OC) and elemental (EC) carbon scavenging efficiencies were determined based on ambient and interstitial aerosol concentrations. The data show that fogs process very efficiently carbonaceous material as up to 85% of the total fine particle carbon was scavenged. Organic carbon was more efficiently scavenged by the studied fog episodes than elemental carbon, which remains mainly in the interstitial particles. This should lead to significant difference in atmospheric lifetimes of organic and elemental carbon in fog prone areas. Scavenging efficiencies in these radiations fogs appear to be correlated to liquid water content with both probably correlated to fog supersaturation, which cannot be measured directly.

Significant differences were observed in scavenging efficiencies between various individual organic compounds. Wood smoke markers including levoglucosan showed very high scavenging efficiencies while other marker compounds like hopanes only weakly interacted with the fogs. This suggests that fogs process organic carbon from different source types with different efficiencies. As fog deposition velocities are high compared to dry deposition velocities, this should lead ultimately to a discrimination of carbonaceous aerosol particles lifetimes according to source type in areas where fog/cloud processing is an important process.

ACKNOWLEDGEMENTS

We are grateful to D. Sherman, J. Reilly, H. Chang and G. Kang for assistance with the CRPAQS fog field campaign and to C. McDade for excellent logistical support. We are grateful to A. Simpson for assistance with the Fresno field campaign and to C. Krauter for hosting the measurements at CSU Fresno. Support for this work was provided by the San Joaquin Valleywide Air Pollution Study Agency and the National Science Foundation (ATM-9980540 and ATM-0222607). The statements and conclusions in this manuscript are those of the Contractor and not necessarily those of the California Air Resources Board, the San Joaquin Valleywide Air Pollution Study Agency, or its Policy Committee, their employees or their members. The mention of commercial products, their source, or their use in connection with material reported herein is not to be construed as actual or implied endorsement of such products.

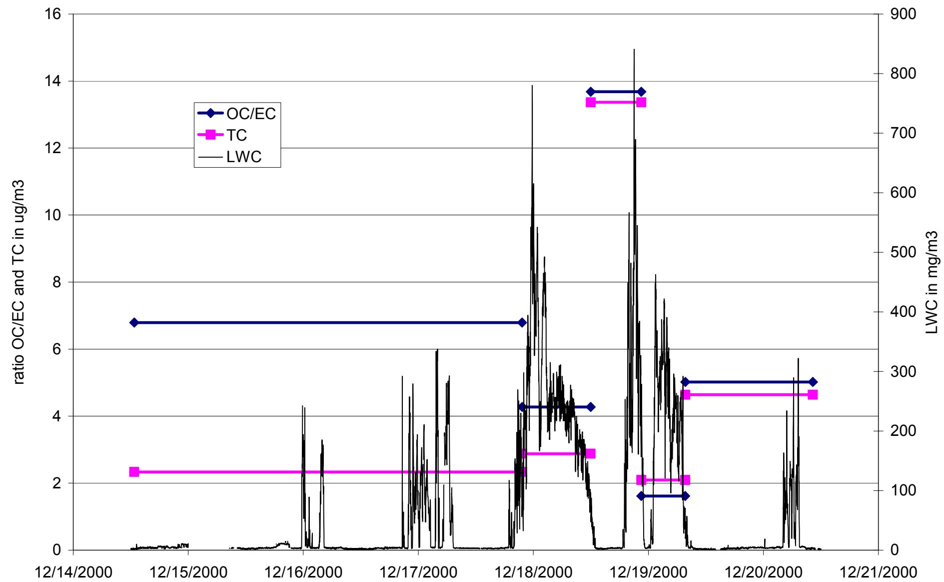

Figure 1. Liquid Water Content (LWC), Total Carbon (TC) and organic to elemental carbon ratio (OC/EC) for Angila (CA) for the period December 14 to December 21, 2000.

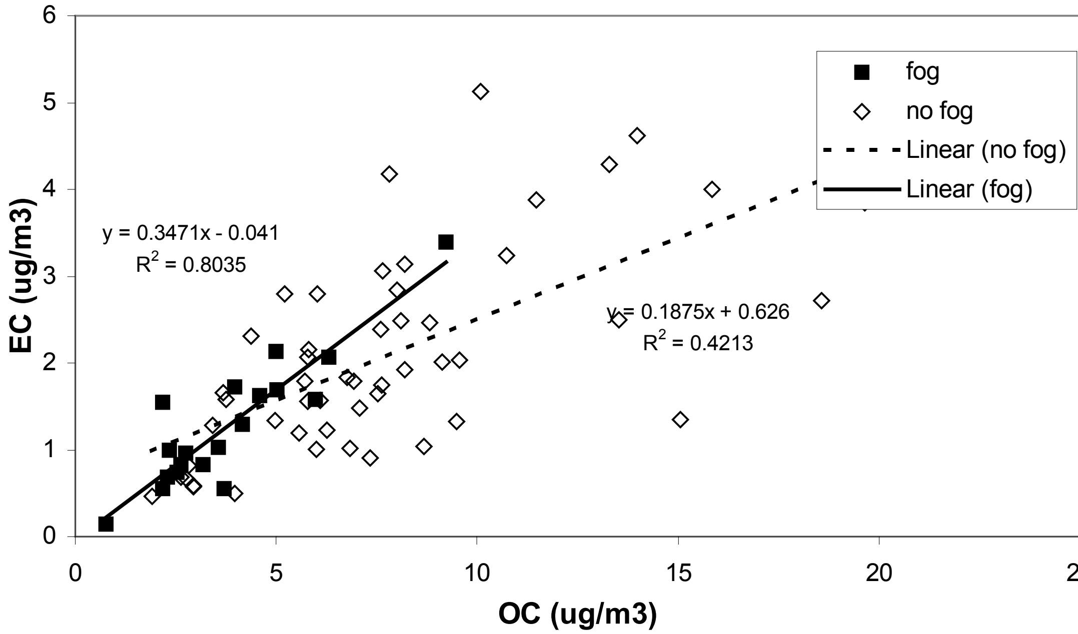

Figure 2. PM2.5 filter concentrations of elemental (EC) vs organic (OC) carbon concentrations as measured during foggy and clear (no fog) periods by Desert Research Institute Scientists at the Angila (CA) site during the CRPQAS winter intensive.

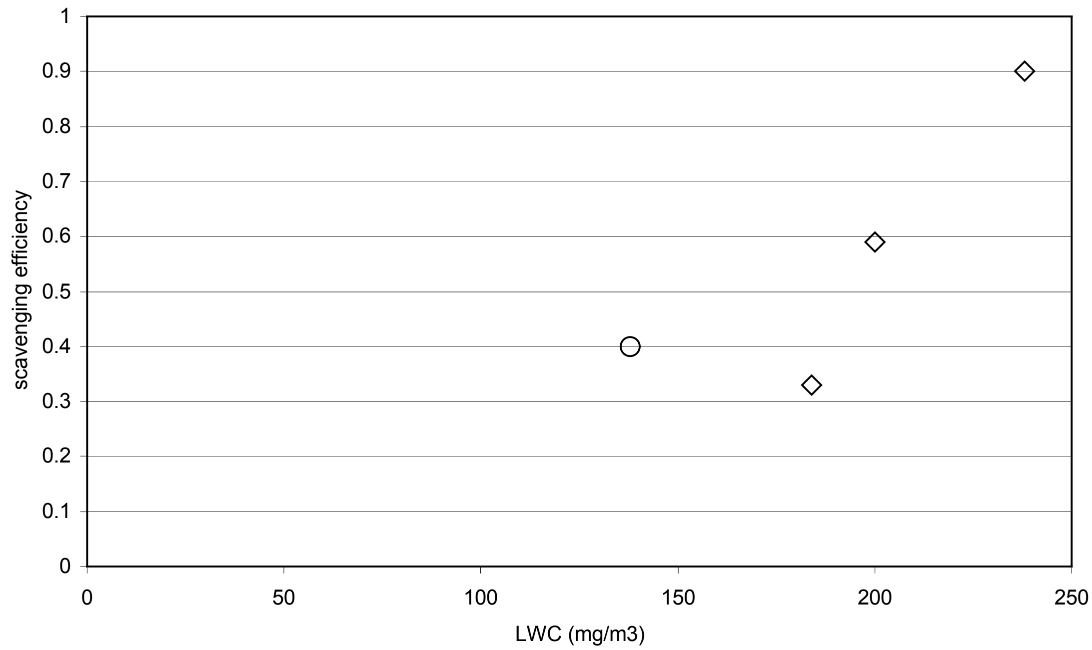

Figure 3. Total Carbon (TC) Scavenging efficiencies as a function of Liquid Water Content (LWC), circles represent data from Fresno, diamonds represent data from Angiola.

Table 1. Scavenging efficiencies η for organic elemental and total carbon

	ηOC^{1}	ηEC^{1}	ηTC^{1}
Angiola 12/19/00	0.90	0.12	0.84
Angiola 1/15/01	0.59	0.05	0.54
Angiola 1/17/01	0.33	0.06	0.29
Fresno 1/11/04	0.44	0^{2}	0.36
Fresno 1/11/04		0.20 (BC)	
Fresno 1/11/04			0.46 (R&P)

1 – TC, OC, EC as defined by the TOT method
2 - no significant difference between pre and interstitial EC concentration

Table 2. Scavenging efficiencies as observed in Angiola (Dec 19/20, 2000) and Fresno (Jan. 11, 2004)

	Angiola	Fresno		Angiola	Fresno
Organic carbon	0.90	0.44	**Elemental Carbon**	0.12	0.00
n-alkanes			***n-alkanoic acids***		
n-C21	0.57	n.d.	**n-C20**	0.67	n.d.
n-C22	0.56	0.69	**n-C21**	0.82	n.d.
n-C23	0.58	0.41	**n-C22**	0.76	n.d.
n-C24	0.61	0.50	**n-C23**	0.79	n.d.
n-C25	0.64	0.58	**n-C24**	0.69	n.d.
n-C26	0.61	0.69	**n-C25**	0.72	n.d.
n-C27	0.63	0.66	**n-C26**	0.79	n.d.
n-C28	0.62	0.74	**n-C27**	0.85	n.d.
n-C29	0.74	0.71	**n-C28**	0.87	n.d.
n-C30	0.55	0.77	**n-C29**	>0.80	n.d.
n-C31	0.73	0.50	**n-C30**	>0.80	n.d.
n-C32	0.34	0.71	**n-C31**	>0.80	n.d.
n-C33	0.66	0.40	**n-C32**	>0.80	n.d.
Average	0.60	0.61			
PAH			**Dicarboxylic acids**		
Fluoranthene	0.59	n.d.	**Adipic acid (C6)**	>0.97	n.d.
Pyrene	0.55	n.d.	**Pimelic acid (C7)**	>0.97	n.d.
benzo[ghi]fluoranthene	0.78	n.d.	**Suberic acid (C8)**	>0.97	n.d.
PAH 252	0.86	n.d.	**Azelaic acid (C9)**	>0.86	n.d.
			Sebacic acid (C10)	>0.86	n.d.
Oxy-PAH					
9,10 – Anthracenedione	>0.85	n.d.			
Benzanthrone	>0.94	n.d.			
Other					`
Syringaldehyde	n.d.	0.88	**Cholesterol**	n.d.	0.67
Vanillin	n.d.	0.96	**Diazinon**	n.d.	0.56
Retene	n.d.	0.63	**17a21b hopane**	0.33	0.68
Levoglucosan	>95%	0.95			

n.d. non determined

>X, not etected in the interstitial particles, scavenging efficiencies calculated based on detection limit

References

Anastasio, C., Faust, B.C., Allen, J.M., 1994. Aqueous-Phase Photochemical Formation of Hydrogen-Peroxide in Authentic Cloud Waters. Journal of Geophysical Research-Atmospheres, 99, 8231-8248.

Birch, M.E., Cary, R.A., 1996. Elemental carbon-based method for monitoring occupational exposures to particulate diesel exhaust. Aerosol Science and Technology, 25, 221-241.

Blando, J.D., Turpin, B.J., 2000. Secondary organic aerosol formation in cloud and fog droplets: a literature evaluation of plausibility. Atmospheric Environment, 34, 1623 - 1632.

Brown, S., Herckes, P., Ashbaugh, L., Hannigan, M.P., Kreidenweis S.M., Collett Jr.,J.L., 2002, Characterization of organic aerosol present at Big Bend National Park, Texas during the Big Bend Regional Aerosol and Visibility Observational (BRAVO) Study, Atmospheric Environment, 36, 5807-5818.

Collett Jr. J.L., Sherman, D.E., Moore, K., Hannigan, M.P., Lee, T., 2001. Aerosol paticle processing and removal by fogs: observations in chemically heterogeneous Central California radiation fogs. Water Air and Soil Pollution: Focus, 1, 303-312.

Corrigan, C.E., Novakov, T., 1999. Cloud condensation nucleus activity of organic compounds: a laboratory study. Atmospheric Environment, 33, 2661-2668.

Cruz, C.N., Pandis, S.N., 1997. A study of the ability of pure secondary organic aerosol to act as cloud condensation nuclei. Atmospheric Environment, 31, 2205-2214.

Demoz, B., Collett, Jr., J. L., Daube, Jr., B. C., 1996. On the Caltech Active Strand Cloudwater Collectors. Atmospheric Research, 41, 47-62.

Ervens, B. Herckes, P., Feingold, G., Lee, T., Collett, Jr., J.L., Kreidenweis, S.M., 2003. On the drop-size dependence of organic acid and formaldehyde concentrations in fog, Journal of Atmospheric Chemistry, 46, 239-269.

Facchini, M. C.; Fuzzi, S.; Lind, J. A.; Fierlingeroberlinninger, H.; Kalina, M.; Puxbaum, H.; Winiwarter, W.; Arends, B. G.; Wobrock, W.; Jaeschke, W.; Berner, A.; Kruisz, C.. 1992. Phase-Partitioning and Chemical-Reactions of Low-Molecular-Weight Organic-Compounds in Fog. Tellus Series B-Chemical and Physical Meteorology, 44, 533-544.

Facchini, M. C.; Fuzzi, S.; Zappoli, S.; Andracchio, A.; Gelencser, A.; Kiss, G.; Krivacsy, Z.; Meszaros, E.; Hansson, H. C.; Alsberg, T.; Zebuhr, Y. 1999. Partitioning of the organic aerosol component between fog droplets and interstitial air. Journal of Geophysical Research-Atmospheres, 104, 26821-26832.

Fahey, K. M., Pandis, S. N., Collett, Jr., J.L., Herckes, P., The Influence of Size-Dependent Droplet Composition on Pollutant Processing by San Joaquin Valley Fogs, Atmospheric Environment, in press

Fuzzi, S. ,Zappoli, S., 1996. The organic component of fog droplets, 12th International Conference on Clouds and Precipitation, Zurich, Switzerland, pp. 1077-1079.

Gelencser, A., Sallai, M., Krivacsy, Z., Kiss, G., Meszaros, E., 2000. Voltammetric evidence for the presence of humic-like substances in fog water. Atmospheric Research, 54, 157-165.

Gerber, H., 1991. Direct measurement of suspended particulate volume concentration and far-infrared extinction coefficient with a laser-diffraction instrument. Applied Optics, 30, 4824 - 4831.

Hallberg, A.; Ogren, J. A.; Noone, K. J.; Heintzenberg, J.; Berner, A.; Solly, I.; Kruisz, C.; Reischl, G.; Fuzzi, S.; Facchini, M. C.; Hansson, H. C.; Wiedensohler, A.; Svenningsson, I. B. 1992. Phase Partitioning for Different Aerosol Species in Fog. Tellus Series B-Chemical and Physical Meteorology, 44, 545-555.

Hansen, A.D.A., Rosen, H., Novakov, T., 1984. The aethalometer — An instrument for the real-time measurement of optical absorption by aerosol particles. The Science of the Total Environment, 36, 191-196.

Herckes, P., Mirabel, P., Wortham, H., 2002a. Cloud water deposition at a high-elevation site in the Vosges Mountains (France). Science of the Total Environment, 296, 59-75.

Herckes, P., Lee, T., Trenary, L., Kang, G.U., Chang, H., Collett Jr., J.L., 2002b. Organic matter in Central California radiation fogs. Environmental Science & Technology, 36, 4777-4782.

Herckes, P., Hannigan, M.P., Trenary, L., Lee, T., Collett Jr., J.L., 2002c. Organic compounds in radiation fogs in Davis (California). Atmospheric Research, 64, 99-108.

Herckes, P., Chang, H., Lee, T., Collett Jr., J.L., An Overview of California Regional Particulate Air Quality Study Fog Episodes and their Effects on Aerosol Formation and Removal, submitted to Journal of the Air and Waste Management Association

Hitzenberger, R.; Berner, A.; Kromp, R.; Kasper-Giebl, A.; Limbeck, A.; Tscherwenka, W.; Puxbaum, H. 2000. Black carbon and other species at a high-elevation European site (Mount Sonnblick, 3106 m, Austria): Concentrations and scavenging efficiencies. Journal of Geophysical Research-Atmospheres, 105, 24637-24645.

Hitzenberger, R.; Berner, A.; Glebl, H.; Drobesch, K.; Kasper-Giebl, A.; Loeflund, M.; Urban, H.; Puxbaum, H.. 2001. Black carbon (BC) in alpine aerosols and cloud water - concentrations and scavenging efficiencies. Atmospheric Environment, 35, 5135-5141.

Laj, P.; Fuzzi, S.; Facchini, M. C.; Lind, J. A.; Orsi, G.; Preiss, M.; Maser, R.; Jaeschke, W.; Seyffer, E.; Helas, G.; Acker, K.; Wieprecht, W.; Moller, D.; Arends, B. G.; Mols, J. J.; Colvile, R. N.; Gallagher, M. W.; Beswick, K. M.; Hargreaves, K. J.; StoretonWest, R. L.; Sutton, M. A. 1997. Cloud processing of soluble gases. Atmospheric Environment, 31, 2589-2598.

Lim, H. J.; Turpin, B. J.; Russell, L. M.; Bates, T. S., Organic and elemental carbon measurements during ACE-Asia suggest a longer atmospheric lifetime for elemental carbon.2003, Environmental Science and Technology, 37, 3055-3061.

Limbeck, A. and Puxbaum, H., 2000. Dependence of in-cloud scavenging of polar organic aerosol compounds on the water solubility. Journal of Geophysical Research-Atmospheres, 105(D15): 19857-19867.

Loflund, M.; Kasper-Giebl, A.; Schuster, B.; Giebl, H.; Hitzenberger, R.; Puxbaum, H. 2002. Formic, acetic, oxalic, malonic and succinic acid concentrations and their

contribution to organic carbon in cloud water. Atmospheric Environment, 36, 1553-1558.
Prenni, A. J.; DeMott, P. J.; Kreidenweis, S. M.; Sherman, D. E.; Russell, L. M.; Ming, Y. 2001. The effects of low molecular weight dicarboxylic acids on cloud formation. Journal of Physical Chemistry A, 105, 11240-11248.
Prenni, A.J., De Mott, P.J., Kreidenweis, S.M., 2003. Water uptake of internally mixed particles containing ammonium sulfate and dicarboxylic acids. Atmospheric Environment, 37, 4243-4251.
Schauer, J.J., Rogge, W.F., Hildemann, L.M., Mazurek, M.A., Cass, G.R., 1996. Source apportionment of airborne particulate matter using organic compounds as tracers, Atmospheric Environment, 30, 3837-3855.
Sellegri, K.; Laj, P.; Dupuy, R.; Legrand, M.; Preunkert, S.; Putaud, J. P. 2003. Size-dependent scavenging efficiencies of multicomponent atmospheric aerosols in clouds. Journal Of Geophysical Research-Atmospheres, 108, A.rt No 4334.
Simoneit, B. R. T., Schauer, J. J., Nolte, C. G., Oros, D. R., Elias, V. O., Fraser, M. P., Rogge, W. F., Cass, G. R., 1999. Levoglucosan, a tracer for cellulose in biomass burning and atmospheric particles. Atmospheric Environment, 33, 173-182.
Voisin, D., Legrand, M., Chaumerliac, N., 2000. Scavenging of acidic gases (HCOOH, CH3COOH, HNO3, HCl, and SO2) and ammonia in mixed liquid-solid water clouds at the Puy de Dome mountain (France). Journal of Geophysical Research-Atmospheres, 105, 6817-6835.
Yu, S.C., 2000. Role of organic acids (formic, acetic, pyruvic and oxalic) in the formation of cloud condensation nuclei (CCN): a review. Atmospheric Research, 53, 185-217.
Zhang, Q., Anastasio, C., 2001. Chemistry of fog waters in California's Central Valley - Part 3: concentrations and speciation of organic and inorganic nitrogen. Atmospheric Environment, 35, 5629-5643.